

# Operator Exposure When Applying Amenity Herbicides by All-Terrain Vehicles and Controlled Droplet Applicators

P. D. JOHNSON<sup>1\*</sup>, D. A. RIMMER<sup>1</sup>, A. N. I. GARROD<sup>2</sup>, J. E. HELPS<sup>3</sup> and C. MAWDSLEY<sup>2</sup>

<sup>1</sup>Health and Safety Laboratory, Harper Hill, Buxton, Derbyshire, SK17 9JN, UK; <sup>2</sup>Health and Safety Executive, Magdalen House, Stanley Precinct, Bootle L20 3QZ, UK; <sup>3</sup>Health and Safety Executive, Government Buildings, Ty Glas, Llanishen, Cardiff CF14 5SH, UK

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A total of 33 surveys of amenity herbicides took place during 1998–1999. These surveys concentrated on two application methods: all-terrain vehicles (ATVs) and controlled droplet applicators (CDAs). The purpose of these surveys was to measure surface deposition and potential inhalation exposure of the operators to the spray fluid used. This paper recommends that the following indicative values should inform risk assessments for these types of application.

ATV: The potential dermal exposure (PDE) to spray fluid (21 sample sets) ranged between 0.7 and 6.8 ml/h of spray fluid, median 2.0 ml/h based on patch samplers. Exposure to the hands, as collected on cotton gloves, ranged between 0.6 and 13.6 ml/h, median 3.0 ml/h. Potential exposure to spray fluid by inhalation was found in 85% of the samples, range 7–37 mg/m<sup>3</sup>, median of non-zero values at 16 mg/m<sup>3</sup>.

CDA: The PDE to spray fluid (12 sample sets) ranged between 0.003 and 0.826 ml/h of fluid, median 0.133 ml/h, based on patch samplers. Exposure to the hands, as collected on cotton gloves inside protective gloves, ranged up to 0.06 ml/h, median 0.004 ml/h, and on socks ranged up to 0.05 ml/h, median 0.001 ml/h. Potential exposure by inhalation was low: detected in just 33% of the samples, range 0.02–0.61 mg/m<sup>3</sup>, median 0.12 mg/m<sup>3</sup>.

Keywords: herbicides; pesticides; exposure; all-terrain vehicle; controlled droplet applicator

## INTRODUCTION

One of the functions of city and county councils in the UK is the control of weeds in and around residential areas as well as on the roadside. Weed growth causes many problems. It can interfere with visibility for road users, obscure traffic signs, block drains and may even damage walls, kerbs and paved or tarmac surfaces. It is therefore important that weed growth in public areas is managed. The most common approach to weed control is the application of herbicides, usually by spraying. Herbicides are typically applied to weeds on road verges, pavement edges, fence and wall lines, brick-paved areas, around utility poles, along walls or fences and in cracks between paving slabs. Each area may be treated

two or three times a year usually in the spring and/or summer, with additional applications later in the year if required.

Herbicides, like other pesticides, have the potential to cause harm to those who apply the products and indirectly to other people, animals, plants and the environment. An assessment of the risks in using pesticides part of the process of placing them on the market. Assessing the risks to humans involves comparing the potential exposure during use with the amounts at which no effects are seen in animal toxicological studies, modified by suitable extrapolation factors to account for differences between and within species.

The Health and Safety Executive (HSE) has been asked for its opinion on the risks to the public from the use of herbicides on amenities, but there is little or no information in the public domain. To inform responses and subsequent risk assessment, research was commissioned to assess the potential inhalation and

\*Author to whom correspondence should be addressed:  
Tel: +44 114 289 2706; fax: +44 114 289 2362;  
e-mail: paul.johnson@hsl.gov.uk

dermal exposure of operators, and also to collect information on the patterns of use and the application equipment. To this end two surveys were carried out using two types of spraying equipment: all-terrain vehicles (ATVs) with front-mounted spray bars and a separate spray lance, and knapsack sprayers equipped with controlled droplet applicators (CDAs).

The study populations were a team of six spray operators for ATV applications and six teams of two sprayers for CDA applications, all of whom were employed by the Local Authorities (LA). Because a significant route for exposure to herbicides is the skin, surface deposition data were needed as well as personal air sampling. It was necessary to know the exact concentration of the pesticide being sprayed and so bulk samples of the spray fluids were also collected.

Both of these surveys involved the use of glyphosate, which is one of the most widely used herbicides in weed control. Glyphosate is a broad-spectrum, non-selective, post-emergence herbicide. It is registered for a variety of agricultural and non-agricultural applications including weed control in non-crop areas, e.g. roadsides, pathways and gardens.

## SURVEYS

The aim of these surveys was to acquire data through sampling in real work situations, to improve knowledge on the range of exposures in applying amenity herbicides and to report occupational hygiene findings in this field of work. HSE and Health and Safety Laboratory (HSL) staff carried out the exposure monitoring and analytical work during 1998 and 1999.

### ATV survey

The first survey took place in the north-west of England in 1998. The traditional method of applying herbicides was by knapsack sprayers, which is labour-intensive, requiring frequent mixing and loading episodes, with associated risks of musculoskeletal disorders. The Council hired six ATVs fitted with rear-mounted pesticide tanks and front-mounted spray bars. The aim was for Council employees to cover over 2800 km of streets within a 6–8 week cycle, starting from mid May, within constraints imposed by the weather.

The herbicide chosen was 'Roundup Pro Biactive', containing 360 g/l glyphosate acid as the isopropylamine salt. The dilution was 5 l of concentrate in 70 l of water—the maximum load of the ATVs—a nominal concentration of 25 g/l of glyphosate. The tank mix was prepared by half-filling with water, adding concentrate and then filling to 70 l using tap water. Each tank load would take around 30 min to discharge, using the front-mounted sprayers while

Table 1. ATV survey summary

Depot	Sample set and operator identification			
	Pad method			Whole-suit method
A	1 (A)	2 (B)	3 (C)	2 (B)
A	4 (A)	5 (B)	6 (C)	4 (A)
B	7 (D)	8 (E)		8 (E)
A	9 (A)	10 (B)	11 (C)	9 (A)
B	12 (D)	13 (E)	14 (F)	12 (D)
B	15 (D)	16 (E)	17 (F)	17 (F)
B	18 (F)	19 (D)		18 (F)
B	20 (F)	21 (D)		

The numbers 1–21 represent the sample sets and the bracketed letters indicate which of the six operators was involved.

moving and a hand-lance for stationary applications in confined places, e.g. around lamp posts. The ATV forward speed was no greater than 7 km/h. The measurement of exposure involved the driver of the ATV using the front-mounted sprayers only and did not include the use of the lances. Sampling time was approximately 30 min, i.e. the time taken for one tank load of herbicide to be used. The mixing and loading activities were completed before the sampling equipment was deployed on the operators. A sample set was taken for each task, i.e. if the operator used two tank loads then the samplers were replaced with fresh ones prior to the second spraying. Eight surveys were conducted involving six operators taking 21 sample sets, working out of two depots. This survey is summarized in Table 1.

During routine spraying work, LA provided their operators with disposable coveralls and protective nitrile gloves. The coveralls were normally worn at all times until they fell apart but the nitrile gloves were usually not worn while driving the ATV. The results for HSL cotton glove samplers (see 'Sampling methods') therefore represent actual hand exposure during driving. Drivers did not normally wear head protection, but for the sampling exercise they wore HSL supplied caps with pad samplers (see 'Sampling methods'). All the drivers were conscious of the safety risks to the general public when driving ATVs on pavements and access routes. The survey work was severely hampered by high winds, rain-storms or both, and took numerous attempts to get started. However the Council employees cooperated well and the survey work was conducted in May 1998.

### CDA survey

A second survey was carried out in Scotland in 1999. The aim was to measure exposure arising from spraying at ground level using CDA sprayers, to note factors that could influence exposure and to record usage patterns. Five LA agreed to cooperate with the survey. Herbicide was to be applied to weeds in typical areas as listed in the 'Introduction'.

Treatment was conducted between May and October, weather permitting. Each area was to be treated two or three times a year. The employees would rotate jobs and so exposure would not occur every day.

CDA sprayers are lances with lightweight spinning discs, fed with pesticide from a knapsack container. The lance handle contains a trigger, flow controls and a battery power source. The controllable variables are the pesticide flow rate, the disc spinning speed (which relates to the aerosol particle size generated and the distance thrown) and the operator's walking speed. Different discs give different spray patterns, a 'spiral' disc gave a spray swath of 10–30 cm, a 'square' disc gave 5–60 cm and a 'serrated' disc gave 60–90 cm. In these surveys, since narrow spray widths were required, 'spiral' and 'square' discs were generally used. Operators held their lances in front of them, about 15 cm above the ground, directing the spray at the area for treatment. On paved areas, the operators swung the lances from side to side, walking forward into the spray aerosol. For road verges, they moved parallel with the verge, walking sideways. Two types of CDA sprayers were used, the NOMIX SUPERPRO 800 was used by 10 of the operators while the other two operators used the EASY MARK LANDSCAPER.

All the products used glyphosate as the isopropylamine salt and were Total Herbicide (NOMIX) ready-for-use packs (two operators), Hilite Herbicide (NOMIX) ready-for-use packs (four operators) and Roundup Pro Biactive (Monsanto) diluted with 'Lightning' to give a 40% strength solution (six operators). The Roundup Pro Biactive was prepared by adding 2 l of concentrate to a 5 l knapsack container half-filled with 'Lightning' diluent and then topping up to full volume with 'Lightning'. The composition of 'Lightning' has not been ascertained. The CDA survey is summarized in Table 2.

Operators normally wore coveralls and safety boots or wellingtons and most of the operators also wore protective gloves. However, during hot weather this situation can change. Some operators removed coveralls entirely or tied the top half around their waist—two of the operators wore shorts. Disposable coveralls were replaced weekly, and the operators washed any other work clothing at home.

Table 2. CDA survey summary

Depot	Operators	Product used	Treatment area
C	22, 23	Ready for use	Pavements
D	24, 25	Ready for use	Pavements
E	26, 27	Ready for use	Pavements
F	28, 29	Diluted	Verges, edges
G	30, 31	Diluted	Kerbsides, pavements
G	32, 33	Diluted	Verges, edges, fence lines

## SAMPLING METHODS

### Surface sampling

Standard patches were used to collect the potential exposure data. These consisted of seven cotton filiated swabs (10 cm × 10 cm; Philip Harris Medical, Birmingham, UK) and were fitted to the operator's clothing with safety pins in the positions set out in Fig. 1. This approach is based on the standard protocol for exposure to pesticides developed by the WHO (1982) and reported in guidance from the OECD (1997). This sampling approach has been successfully applied in previous HSE surveys (Llewellyn et al., 1996; Garrod et al., 1998). Recommended sampling methods for assessing potential dermal exposure (PDE) have now been published by HSE (EH74/3, 1999; MDHS 94, 1999) and have since been used in further surveys (Garrod et al., 1999, 2000).

The pad in position 3 was placed to give information on the quantity of herbicide getting through the top layer of clothing. To determine hand contamination, lightweight cotton gloves (RS Electrical Components, Corby, UK) were worn; if protective gloves were worn by the operator these were worn over the sampling gloves. For the CDA survey, Wilson cotton sports socks were worn under the operator's boots to determine foot exposure. The patch sampling method has been validated for spray applications (Tannahill, 1996; Glass, 1998). In addition to patch samples, Tyvek<sup>1</sup> semi-absorbent disposable suits were worn by seven of the operators in the ATV survey to draw a

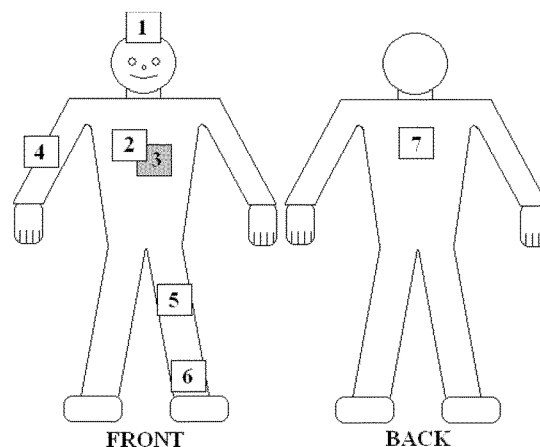


Fig. 1. Position of the seven sampling pads used in the WHO sampling protocol. Position 1: on the hat, as close as practicable to the top of the head. Position 2: over the sternum, on the outside of normal clothing. Position 3: on the sternum, on the inside of normal clothing. Position 4: upper surface of the right forearm held with the elbow bent at right angles across the body, midway between elbow and wrist, on the outside of normal clothing. Position 5: front of left leg, mid-thigh, on the outside of normal clothing. Position 6: front of left leg, above the ankle, on the outside of normal clothing. Position 7: on the back between shoulder blades, on the outside of normal clothing.

comparison of the patch sampling method with the whole-suit approach. This was carried out at the same time as the patch sampling.

#### Potential exposure by air

Air at 0.5 l/min was drawn through a glass fibre (GF/A) filter (Whatman, Maidstone, UK), held in a modified UKAEA sampling head with a standard 45 mg Tenax<sup>1</sup> sorbent tubes (SKC, Dorset, UK, part no. 226-35) downstream of the filter, mounted in the breathing zone of the operator, on the left shoulder (MDHS 94, 1999).

#### Blank samples

Blank samples of surface and inhalation samplers were taken while on site and treated in the same way as the personal samples.

#### Bulk samples

Bulk samples (10 ml) of concentrates and, where appropriate, diluted working solutions were taken in plastic bottles. To prevent cross-contamination, all sampling media were packed in separate plastic bags and stored below 4°C.

#### Analytical procedures

Glyphosate (97.5%, Qm<sub>x</sub> Laboratories Ltd, UK) was prepared gravimetrically as a 100 mg/ml solution in deionized water (Milli-Q Plus Water purifier, 18 M Vcm). Five working standards were prepared in the range 50–1000 ng/ml by derivatizing 50 ml of appropriately diluted stock as described below. All standards and stock solutions were stored in amber bottles at 2–8°C. Citral (3,7-dimethyl-2,6-octadienal, 95%, Aldrich Chemical Co., UK) was diluted with ethyl acetate (Distol grade, Fisher Scientific, UK) to give a 0.2% solution. The derivatization mixture (Aldrich) of trifluoroacetic anhydride (TFAA) and 2,2,3,3,4,4,4-heptafluoro-1-butanol (HFBA) (2:1) was freshly prepared and cooled (#20°C).

Blank devices, six of each, were fortified at 50 mg glyphosate per pad, 200 mg per glove, 400 mg per sock and 4 mg per Tenax tube and GF/A filter. The samples and spikes were extracted into appropriate amounts of water, 25 ml for pads, 100 ml for gloves, 200 ml for socks, 1500 ml for wholesuits and 2 ml for the Tenax tubes and GF/A filters. The samples were then shaken vigorously (1 min) and placed in an ultrasonic bath (30 min). A small aliquot (1–2 ml) was passed through a syringe filter and then an aliquot of 50 ml was transferred to a silanized Reacti-Vial (10 ml, Perbio science Ltd, UK) ready for derivatization. The working solutions and concentrates were also analysed by this procedure after appropriate dilution in water.

The derivatization mixture (1.5 ml) was added slowly to the sample in the Reacti-Vial. The vial was then sealed and the reaction mix was heated to

Table 3. Glyphosate recovery data ( $-1 \cdot \text{SD}$ ,  $n = 6$ ) and estimated limits of detection ( $3 \cdot \text{S/N}$ ) from spiking experiments

Device	1998 results	1999 (CDA) results (%)	LOD
Pad	104 – 7%	113 – 4	0.3 mg/device
Glove	99 – 9%	117 – 16	4 mg/device
Sock	Not performed	109 – 12	2 mg/device
GFA	105 – 6%	104 – 11	0.2 mg/m <sup>3</sup>
Tenax	Not performed	90 – 12	0.2 mg/m <sup>3</sup>

95°C for 2 h in heating blocks to allow derivatization. After cooling, the excess reagents were removed under a gentle stream of nitrogen using sample concentrators set at 40°C. Ethyl acetate/citral (200 ml) was then added to dissolve the samples. The solutions were then transferred to GC vials (250 ml microvials) and capped, ready for analysis.

The analysis was performed on a Hewlett Packard 6890 gas chromatograph fitted with a Hewlett Packard HP-5 MS column (cross-linked 5% phenyl silicone, 30 m · 2.5 mm · 2.5 mm film thickness) and a Hewlett Packard 5973 Series Mass Selective Detector with Hewlett Packard G1701BA MS ChemStation software. The injection (splitless) and transfer line temperatures were 250 and 280°C, respectively, and the injection volume was 1 ml. The oven temperature programme was 60°C for 1 min, ramping at 20°C/min to 240°C; the total run time was 10 min. Helium (>99.996%) was used as the carrier gas and electronic pressure control in constant flow mode delivered 0.98 ml/min. Selected ion monitoring (SIM) data was collected between 5.0 and 7.5 min. In electron impact mode the SIM ion was  $m/z = 612$  for glyphosate.

The limits of detection (LOD) were determined from three times the signal to noise ratio (S/N) calculated from a macro in the HP G1034C MS ChemStation software for the lowest standard. The analytical recoveries and LOD, which are given in Table 3, were considered satisfactory (EU, 2000) for this type of work. Full details of the analytical method can be found in a separate report (Johnson et al., 2003).

## RESULTS

#### Calculation of potential exposure

The standard patches (i.e. 10 cm · 10 cm) and the bulk samples were analysed for the active ingredient, glyphosate. From this information the deposition of spray fluid on the coveralls (PDE) was estimated. The amount of active ingredient on each sampling pad was divided by the concentration of the spray solution to give the amount of spray solution on each pad. This was then multiplied by a correction factor to give the

Table 4. Calculation of deposition of spray fluid on coveralls

Sample pad	Body area (cm <sup>2</sup> )	% Area covered by pad	Multiplication factor
Head	1450	6.9	14.5
Chest	4620	2.2	46.2
Arms	2020	4.9	20.2
Upper legs	3640	2.7	36.4
Lower legs	3640	2.7	36.4
Back	4620	2.2	46.2

body part equivalent. Table 4 lists the area correction factors used. The potential exposure was then calculated by summing the body part equivalents. The combined results were then divided by two to allow for the unevenness of deposition in the areas where direct exposure was less likely, e.g. creases in the overalls, the insides of legs and under the arms. This approach has been supported by work carried out at the Central Science Laboratory, UK (Glass, 1998). The resultant PDE data are expressed in terms of the quantity of in-use spray fluid per hour.

The deposition rates were determined by dividing the amount of spray fluid deposited on coveralls by the job time. The hand and foot exposures are reported as the volume of spray fluid per hour and the potential exposure by inhalation as a time-weighted average over the sampling period. The results from the Tenax and GF/A samplers were summed to give a total value for vapour and aerosol. No corrections for recovery were made.

#### Data

The results for each sample set are summarized in Tables 5 (ATV survey) and 6 (CDA survey). The glyphosate concentrations of the in-use spray fluids for the surveys are shown in each table. Samples that produced undetected results have been assigned a zero value.

The results of the survey appear in Tables 7 (ATV survey) and 8 (CDA survey) and are expressed in a general form 'ml/h spray fluid' such that they may be applied to spraying any fluid for any time period in tasks that are similar to those reported in this survey. The potential inhalation data are reported in a general form mg/m<sup>3</sup>.

The pattern of deposition on clothing is reported in Tables 9 (ATV survey) and 10 (CDA survey). This shows where the spray fluid is deposited on the various body surfaces. The values are given as averaged and normalized ratios.

Some of the ATV sprayer operators wore Tyvek suits in addition to the pad samplers. These were analysed separately to determine the correlation between the two sampling methods. The results are shown in Table 11.

Table 5. Potential dermal, hand and inhalation exposure to the spray fluid for the ATV survey

Data	Job time (min)	Spray fluid				
		'In-use' (g/l)	PDE (ml/h)	Hands (ml/h)	Inhalation (mg/m <sup>3</sup> )	Suit (ml/h)
1	35	36	4.2	6.3	36.5	
2	30	39	1.9	2.9	10.2	7.4
3	40	34	2.5	1.2	9.0	
4	45	23	2.5	5.1	34.6	7.5
5	40	46	6.0	7.3	6.5	
6	45	27	2.0	3.0	0	
7	30	31	6.8	5.3	12.9	
8	25	33	1.7	2.5	14.5	2.2
9	30	55	2.6	2.3	7.2	1.1
10	40	25	0.8	3.4	12.1	
11	40	34	0.7	0.9	0	
12	40	18	1.2	3.9	16.6	7.2
13	35	21	1.7	8.1	16.7	
14	30	20	3.7	2.2	20.0	
15	30	11	4.0	13.6	35.4	
16	25	43	1.5	2.3	11.2	
17	30	17	1.4	3.3	23.0	3.4
18	25	24	2.6	2.1	0	10.4
19	30	22	1.8	3.6	18.3	
20	30	20	2.0	0.6	20.4	
21	35	36	0.8	0.8	9.8	

Table 6. Potential dermal, hand, foot and inhalation exposure for the CDA survey

Data	Job time (min)	Spray Fluid				
		'In-use' (g/l)	PDE (ml/h)	Hands (ml/h)	Feet (ml/h)	Inhaled (mg/m <sup>3</sup> )
22	164	126	0.009	0.003	0.0013	0
23	171	126	0.175	0.004	0.0005	0
24	155	72	0.159	0.004	0.0011	0
25	155	72	0.017	0.008	0.0014	0
26	100	78	0.014	0.001	0.0013	0
27	100	78	0.050	0.010	0.0161	0
28	170	99	0.826	0.060	0.0455	0.616
29	170	99	0.678	0.007	0.0105	0.222
30	271	167	0.106	0.002	0.0007	0.022
31	228	167	0.003	0.001	0.0001	0.026
32	191	167	0.440	0.003	0.0002	0
33	191	167	0.666	0.001	0.0001	0

Table 7. Exposure ranges and indicative values for the ATV survey

Value	PDE <sup>a</sup> (ml/h)	Potential hand exposure (ml/h)	Inhalation exposure (mg/m <sup>3</sup> )
Non-zero	21	21	18
Range	0.7–6.8	0.6–13.6	6.51–36.5
Median values	2.0	3.0	15.6
75th percentile	2.6	5.1	20.2
95th percentile	6.0	8.1	35.5

<sup>a</sup>PDE data excludes whole-suit data.

Table 8. Exposure ranges and indicative values for the CDA survey

Value	PDE (ml/h)	Potential hand (ml/h)	Potential foot (ml/h)	Inhalation (mg/m <sup>3</sup> )
Ready for use (n = 6)				
Non-zero	6	6	6	0
Range	0.009–0.175	0.001–0.010	0.0005–0.0161	0
Median	0.034	0.004	0.001	0
75th percentile	0.132	0.007	0.001	0
95th percentile	0.171	0.010	0.012	0
Diluted (n = 6)				
Non-zero	6	6	6	4
Range	0.003–0.826	0.001–0.060	0.0001–0.0455	0.02–0.62
Median	0.553	0.003	0.001	0.12
75th percentile	11.25	0.10	0.13	0.32
95th percentile	13.15	0.76	0.61	0.56
All results (n = 12)				
Non-zero	12	12	12	4
Range	0.003–0.826	0.001–0.060	0.0001–0.0455	0.02–0.62
Median	0.133	0.004	0.001	0.12
75th percentile	0.675	0.006	0.008	0.32
95th percentile	0.789	0.047	0.037	0.56

Table 9. Distribution pattern of deposition on coveralls for the ATV survey

	Head	Arms	Chest	Back	Legs
Range (%)	1–89	0.4–48	0–17	1–20	8–93
Median (%)	3.7	3.7	8.3	8.0	67.7
Normalized (%)	4.0	4.0	9.1	8.8	74.1

Table 10. Distribution pattern of deposition on coveralls for the CDA survey

	Head	Arms	Chest	Back	Upper legs	Lower legs
Range (%)	0–4	0–27	0–21	0–86	1–71	0–97
Median (%)	0.2	1.0	1.3	9.5	15.6	51.1
Normalized (%)	0.3	1.3	1.7	12.0	19.8	64.9

Table 11. Comparison of calculated PDE for whole-suit data and pad data

Sample no.	PDE from whole-suit data (ml/h)	PDE from patch data (ml/h)	Ratio
2	7.4	1.9	3.9
4	7.5	2.5	3.0
9	2.2	1.7	1.3
10	1.1	2.6	0.4
13	7.2	1.2	6.0
18	3.4	1.4	2.4
22	10.4	2.6	4.0

## DISCUSSION

Studies quantifying PDE in non-agricultural occupations are sparse, and there are no reported studies relating to amenity spraying. Previous and

subsequent research into PDE (e.g. Llewellyn et al., 1996; Garrod et al., 1998) show the expected variability to be several orders of magnitude greater than is anticipated for inhalation exposure. Given this variability, advanced statistical treatment was not deemed appropriate. The work reported in this paper suggests the ranges of PDE that can be expected in the use of CDA and ATV application of herbicides. It characterizes the magnitudes and ranges of worker exposure in these tasks. The results are intended for use in HSE's role as a regulator.

Exposure via the skin is an important route for exposure during spraying. The main conclusions concern the rates of deposition of spray fluids on the surfaces of workers' coveralls and penetration thereof, on socks and inside gloves. PDE relates to the amount of residue, individual behaviour and the exact nature of the tasks performed. The rates are therefore normalized and expressed as a quantity of spray fluid rather than glyphosate, to make them applicable to any similar spraying operation. Quoting results in this general and normalized form enables predictive and quantitative risk assessments to be made.

## Comparison of application techniques

The deposition rates, i.e. PDE, expressed as the volume of in-use spray fluid per hour were found to be considerably higher for the ATV sprayers than for the CDA spraying. The 21 data sets of PDE for the ATV sprayers ranged between 0.7 and 6.8 ml/h of spray fluid, median 2.0 ml/h based on patch sampling. The 12 data sets for the CDA sprayers ranged between 0.003 and 0.826 ml/h,

median 0.133 ml/h, based on sampling patches. It can be seen that although the actual levels of contamination for the ATV spraying are much higher than for the CDA spraying, the pattern of distribution is similar with the majority of the contamination being on the legs. The split on the legs shows that the majority of the spray is deposited on the lower legs for CDA spraying, about 70%. This would have been expected as the spray is directed down to the ground in front of the operator. For the ATV spraying the distribution of the spray is less obvious, but generally the lower legs are more contaminated than the upper legs. The higher deposition rates may be a consequence of the different ways that the two sprayers operate. With CDA application the spray is directed down with the operator walking forward, while with ATV application the spray is from front-mounted sprayers that are raised above the ground. Given this, it is would be expected that ATV spraying would allow for greater exposure and that CDA spraying would concentrate exposure on the lower legs. This is shown in the data with the median value for the PDE being an order of magnitude higher for ATV spraying than for CDA spraying.

There is only one major difference in the distribution of spray fluid on the torso. For ATV spraying, 50% is found on the front of the torso and 50% on the back, while for the CDA spraying the split is 12% on the front and 88% on the back. This is likely to be due to the herbicide reservoir being carried on the back for CDA spraying, causing a high level of contamination in this area. It is likely that there will be contamination of the outside of the backpack during filling and a subsequent transfer of the spray fluid from the contaminated surface of the backpack. The backpacks were kept in containers/pouches and these would also become contaminated over time. The relatively high values for operators 28 and 29 in the CDA data set are almost certainly attributable to leaking applicators.

The contamination of the head pads is likely to have occurred when operators inadvertently touched their foreheads with contaminated gloves and hands. Operator 28 had much higher levels of contamination than other subjects and his exposure for all body segments was elevated due to a grossly leaking applicator, this needs to be considered throughout the discussion in this section.

The level of penetration through the coveralls, calculated from the amount of spray fluid found on pad 3 (chest, under the coveralls) compared to pad 2 (chest, over the coveralls) was 11% for the ATV survey. No meaningful data were obtained for personal protective equipment (PPE) penetration in the CDA study. Although contamination to the interior sternum pad was recorded for eight of the twelve subjects, the operators were wearing their normal work clothes and it was not recorded if they were

clean or not. As a result it is highly possible that there would have been a transfer of spray fluid from previously contaminated garments and/or through openings such as buttonholes. As such the results for garment penetration cannot be considered fully reliable. It should also be noted that operator 28 in the CDA set unzipped his coverall for part of the sample period which accounted for the high contamination on his inner pad, which was of a similar magnitude to his outer pad.

The seven whole-suit data obtained for the ATV survey ranged between 1.1 and 10.4 ml/h of spray fluid; these values are of the same order of magnitude as the data obtained using patches. There are, however, few data to compare and the variation of potential exposure data using both patch and suit sampling is high. The ratio of the measured glyphosate on the suit compared with the calculated amount from the patch results gave a mean of 3.0, suggesting that the patch sampling is representative, in terms of order of magnitude, albeit undersampling.

For the ATV survey, operative exposure to the hands as collected on cotton gloves was between 0.6 and 13.6 ml/h, median 3.0 ml/h. For the CDA survey, the exposure was up to 0.06 ml/h, median 0.004 ml/h. The operators did not wear protective gloves while driving ATVs and therefore the levels of hand exposure found for the ATV survey are actual and not potential exposure values. This would also explain the much higher exposure values seen for ATV operators than for CDA operators. For the CDA spraying, most of the operators regularly wore protective gloves during the spraying operation. As with their work clothes these may have been contaminated from use during previous applications, and therefore the hand exposure values reported here may not be totally due to penetration or leakage into the gloves during a particular application. Nevertheless, cross-contamination from reused PPE is still a potential source of skin exposure and the low levels of glyphosate found are likely to be due to this cross-contamination from previously used gloves. The highest levels found on gloves for the CDA operators arose from contact between the contaminated outer glove and the glove sample during removal for a work break. This points out another important factor to take into account when considering overall exposure. Even with the appropriate PPE used correctly there are opportunities for exposure when putting on and removing contaminated equipment.

The data for exposure to feet were collected for CDA spraying only and were found to be quite low. The exposure ranged up to 0.5 ml/h, median 0.001 ml/h. The highest levels were found for workers wearing wellington boots. The rate of exposure from CDA applications on hands and feet is similar, again implying that the exposure is due to cross-contamination and not directly from the spray.

The presence of herbicide on the socks also demonstrates a potential source of skin exposure, despite operators wearing work or wellington boots. It was noted that contamination was visible on the boots of all the CDA operators.

Potential exposure by inhalation was found in 85% of the ATV samples (18 data sets) and ranged between 6.51 and 36.5 mg/m<sup>3</sup>, median of non-zero values 15.5 mg/m<sup>3</sup>. For the CDA survey only 33% of the samples gave positive results (4 data sets) and ranged between 0.02 and 0.62 mg/m<sup>3</sup>, median of non-zero values 0.12 mg/m<sup>3</sup>. The amount of spray fluid potentially inhaled was about 130 times higher for ATV application than for CDA. This is again probably because the spray is directed down to the ground during CDA application and it is more likely that the detection of glyphosate is attributed to contamination of the sampling head by splashing rather than aerosol capture. The high levels found with data points 28 and 29 in the CDA set is most likely to have occurred when the operators were adjusting their applicators rather than during spraying.

The high variability of the exposure levels appears to be dependant on the condition of the sprayer. This was most noticeable in the CDA survey where a faulty applicator led to high levels of exposure for operator 28 than for those using correctly calibrated applicators in good condition, where exposure was restricted primarily to the leg region. When problems were encountered with the applicator, exposure was generally elevated for all body regions. The operator had to adjust the applicator which resulted in contamination of the protective gloves and subsequently indirect transfer to other body regions.

Human factors, such as attitude of the operators and operator technique have a significant effect on exposure. Exposure conscious operators tended to

work with more care and minimized contact with the herbicide, which is reflected in the range of the results.

## REFERENCES

- EH 74/3. (1999) Dermal exposure to non-agricultural pesticides—exposure assessment document. London: HSE Books. ISBN 0-7176-1718-1.
- EU. (2000) Quality control procedures for pesticide residues analysis. European Union guideline document no. SANCO/3103/2000.
- Garrod ANI, Guiver R, Rimmer DA. (2000) Potential exposure of amateurs (consumers) through painting wood preservative and anti-foulant preparations. *Ann Occup Hyg*; 44: 421–6.
- Garrod ANI, Martinez M, Pearson J et al. (1999) Exposure to preservatives used in the industrial pre-treatment of timber. *Ann Occup Hyg*; 43: 543–55.
- Garrod ANI, Rimmer DA, Robertshaw L et al. (1998). Occupational exposure through spraying remedial pesticides. *Ann Occup Hyg*; 40: 159–65.
- Glass CR. (1998) Measurement of potential dermal exposure in Greece and Spain with patch and whole body dosimetry techniques. MAFF Chief Scientists Group Project PA 1701. Central Science Laboratory report.
- Johnson PD, Rimmer DA, Corns H et al. (2003) Determination of glyphosate in water, vegetation and occupational hygiene samples, using gas chromatographic analysis with mass selective detection. HSL report number OMS/2003/26.
- Llewellyn DM, Brazier A, Brown R et al. (1996) Occupational exposure to permethrin during its use as a public hygiene insecticide. *Ann Occup Hyg*; 40: 499–509.
- MDHS 94. (1999) Pesticides in air and/or on surfaces. London: HSE Books. ISBN 0-7176-1619-3.
- OECD. (1997) Environmental health and safety publications series on testing and assessment no. 9: Guidance document for the conduct of studies of occupational exposure to the pesticides during agricultural application. OEDE/GD(97)148. Paris: OECD.
- Tannahill SN. (1996) Institute of Occupational Medicine Phase 1 report. HSE Research Project R3228/R46.60.
- WHO Standard Protocol. (1982) Field surveys of exposure to pesticides. Ref. VBC/82.1 Geneva: WHO.